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Effects of Large (0.9 micrometer) Sulfuric Acid Aerosols on Human Pulmonary Function

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Effects of Large (0.9 μm) Sulfuric Acid Aerosols on Human Pulmonary Function

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The effects of sulfuric acid particle concentration (mass/volume) and ambient temperatures on pulmonary function of young male nonsmokers were examined. Subjects (n = 11) thrice repeated a sequence of 20-min exercise (ventilation approximately 30 liters/min) and 20-min sitting rest. Pre- and postexposure pulmonary function tests (forced vital capacity maneuvers, lung volumes, maximum voluntary ventilation, R_{aw}, TGV, and helium–oxygen FVC) were measured. During the exposure and at 2 min after cessation of each exercise period, forced vital capacity tests (air and helium–oxygen) were performed. Each subject was exposed in random order to filtered air or 233, 418, or 939 μg/m^3 particulate sulfuric acid. Mass median diameter (MMD) was 0.90–0.93 μm, and geometric SD was 1.66–1.73. No major changes in pulmonary function related to sulfate were observed in these subjects. The only measurement to show a significant interaction across time (pre–post) and sulfate concentration was $FEV_{1.0}$, which was significantly decreased with exposure to 939 μg/m^3 sulfate. The magnitude of the decrease was not considered to be of physiological significance. These results indicate that, even with the added stress of the increased depth and volume of exercise ventilation, exposure to as much as 1 mg/m^3 of 1 μm sulfuric acid aerosol for 2 hr has little if any effect on standard tests of lung function.

INTRODUCTION

There has been increased interest recently in the sulfur-containing air pollutants, notably particulate sulfates and sulfuric acid, as a result of concern that there may be increased usage of coal and petroleum products with a high sulfur content. In two early studies (Amdur *et al.*, 1952; Sim and Pattle, 1957) subjects were exposed briefly (10–16 min) to sulfuric acid concentrations of 0.35 to 5 mg/m^3 and 4 to 40 mg/m^3 (particle size 1 μm), respectively, and changes in respiratory patterns, coughing, and bronchoconstriction were reported. More recently, Newhouse *et al.* (1978) exposed ten healthy nonsmokers to 1 mg/m^3 of H_2SO_4 mist (0.5 μm) for 2.5 hr (which included 20 min of exercise), but no significant changes in VC, $FEV_{1.0}$, and MMEF were observed after 2 hr of exposure.

Sackner *et al.* (1978) studied normal resting young adults and middle-aged asthmatic subjects who breathed by mouth either sodium chloride or sulfuric acid aerosols (0.1–0.2 μm) for 10 min at concentrations of 10, 100, and 1000 μg/m^3. Neither normal nor asthmatic individuals showed any alterations in pulmonary functions. Avol *et al.* (1979) also investigated the effects on pulmonary functions of intermittently exercising subjects exposed for 2 hr to sulfuric acid (75 μg/m^3, particle size 0.48–0.81 μm). No convincing adverse short-term health effects were observed. Lippman *et al.* (1981) had ten subjects inhale via nasal mask 0.5

μm H_2SO_4 at 0, 100, 300, and 1000 $\mu g/m^3$ for 1 hr. Although no significant changes in respiratory dynamics were observed, bronchial mucociliary clearance was altered. Exposure to sulfate aerosols (ammonium sulfate, ammonium bisulfate, zinc ammonium sulfate, zinc sulfate, and sodium sulfate) (Sackner *et al.*, 1978; Avol *et al.*, 1979) did not induce any significant effects on pulmonary function at concentrations up to 1000 $\mu g/m^3$. Several preliminary reports (Utell *et al.*, 1979; Leikauf *et al.*, 1979) have appeared suggesting either no effects (Leikauf) or significant changes in flow rates and increased bronchial reactivity (Utell). Kerr *et al.* (1981) evaluated respiratory effects of sulfuric acid aerosol in normal subjects exposed for 4 hr to 100 $\mu g/m^3$ H_2SO_4 of particle size 0.1 to 0.3 μm. Their subjects performed two 15-min bouts of exercise during the exposure period. No significant differences in pulmonary function were observed during or after the exposure. Kleinman *et al.* (1981) have examined the effects of a combination 100 $\mu g/m^3$ H_2SO_4 aerosol, 0.37 ppm SO_2, and 0.37 ppm O_3. Although small decrements in pulmonary function occurred (3.7% decrease in $FEV_{1.0}$), the authors suggested that at least 75% of this effect could be attributed to the effect of ozone.

The size of the sulfuric acid aerosol may be responsible for the different results reported. Since larger H_2SO_4 particles may produce a greater effect, we have determined the responses of normal male nonsmokers to a 2-hr exposure to sulfuric acid aerosols of 0.9 μm.

METHODS

Subjects. Eleven adult males served as subjects in this study (Table 1). Two were former smokers who had not smoked for at least 1 year, and the others had never smoked. Six subjects reported some type of "allergy" and one had had "asthma" as a child. Six subjects previously lived in areas known to have high oxidant pollution, one previously lived in an area of sulfur dioxide pollution, and four came from areas of low air pollution. All subjects had lived in a low-pollution area for 3 months prior to the study. Three subjects reported having experienced respiratory symptoms (cough, chest tightness) when previously exposed to high ambient levels of air pollution.

The purpose and procedures of the experiment were explained to the subject and each gave his informed consent to participate.[1] A medical history, a resting 12-lead electrocardiogram, spirometric tests (forced vital capacity, residual volume, and associated parameters), a submaximal exercise test, and a physical examination were used in evaluating and screening each subject.

Experimental design. The subjects were randomly exposed to filtered air (FA) and 223, 418, and 939 $\mu g/m^3$ of sulfuric acid aerosol (0.9 μm). There was an interval of at least 7 days between exposures. Exposures lasted for 2 hr and occurred at the same time of day for each subject. During the exposures, the subject walked on a treadmill at a speed and slope sufficient to raise ventilation

[1] The nature and purpose of the study and the risks involved were explained verbally and given on a written form to each subject prior to his voluntary consent to participate. The protocol and procedures for this study were approved by the Committee on Activities Involving Human Subjects of the University of California, Santa Barbara.

TABLE 1
PHYSICAL CHARACTERISTICS OF SUBJECTS (n = 11)

	Ht (cm)	Wt (kg)	Age (years)	FVC (ml)	Percentage predicted FVC	$FEV_{1.0}$/FVC (%)
Mean	179.8	71.1	20.9	5576	99.7	84.2
Range	(163–187)	(66–790)	(18–25)	(4814–6819)	(81–116)	(73–90)

volume to approximately 30 liters/min (BTPS). The ambient conditions were 22°C and 55% relative humidity.

Protocol. Prior to exposure, pulmonary function tests (forced vital capacity maneuvers (FVC), functional residual capacity (FRC), airway resistance, thoracic gas volume, and maximum voluntary ventilation (MVV)) were performed. Electrocardiograph chest leads were attached and the subject entered the chamber and began to walk for three 20-min periods with a 20-min rest period between each walk. Forced vital capacity tests were performed during each rest period. At the end of the 2-hr period, the subject left the chamber, was examined by a physician, and then repeated the preexposure pulmonary function tests. The subject was interviewed regarding the symptoms he may have experienced in the chamber.

Measurements. Pulmonary function tests were performed in the standing position using a 7- or 14-liter spirometer (W. E. Collins). Thoracic gas volume and airway resistance were measured in a constant-volume body plethysmograph using the method of DuBois *et al.* (1956*a*, *b*). A wedge spirometer (Med-Science) was used for FVC measurements inside the chamber. Both spirometers were interfaced to a laboratory computer (Digital PDP 11-60) for on-line analysis of pulmonary function tests. All volume measurements were corrected to BTPS.

A double-walled acrylic insulated chamber was used for exposures. An airflow of 2.8 m^3/min, providing an air change approximately once every 3–5 min, was generated by a custom-designed air-conditioning unit (Bemco). Incoming air was passed through a Purafil bed (which removes a large number of air contaminants) and particulate filters (including a HEPA filter). The air was cooled slightly by a cooling coil in the air-conditioning unit and then passed through another set of particle filters before entering the chamber via a 0.5-m^3 mixing box. Sulfuric acid particles were generated using a nebulizer which generates particles in the micron-size range (Walters, 1977). To prevent change in aerosol size, the specific gravity of sulfuric acid (1.355) was chosen so that the water vapor pressure of the acid was in equilibrium with the ambient water vapor pressure in the chamber. A stream of dry nitrogen was used as the carrier gas at pressures ranging from 75–150 Torr. The relationship between nebulizer pressure and sulfate concentration was established prior to any human exposures and was used to determine the sulfate concentration during the exposures. In addition, a flame photometric sulfur analyzer was used to monitor sulfur concentration. Because of the high temperature of the analyzer block in this instrument, much of the sulfuric acid signal is lost (Huntzicker *et al.*, 1978) and for this reason the instrument provided only an estimate of sulfate concentration.

Sulfate particles were collected on glass fiber filters (Gelman type AE) prewashed with nitric acid. Samples were collected for 15 min at a flow rate of 25 liters/min. Filters were then placed in distilled water, macerated, and centrifuged. The supernatant was analyzed for sulfate concentration using the AIHL Turbidimetric Barium Sulfate Method (AIHL, 1965).

Particle size was determined using a 10-stage cascade impactor (IBC-Celesco) which uses quartz crystal microbalances as the impaction surfaces and provides a printout which is proportional to the mass deposited on each impaction stage. During periods in which the chamber was empty, there was less than 1 $\mu g/m^3$ of aerosol in the 0.05- to 25-μm range, the size range of the instrument.

The sulfate concentrations taken from filter samples averaged 223, 418, and 939 $\mu g/m^3$. Mass median particle diameter averaged 0.92 μm with an average geometric standard deviation of 1.7 (Table 2).

Data analysis. Data were treated by a two-factor analysis of variance with repeated measures across pollutant concentration and time period within the exposure. When interactions were significant, a test for simple main effects was applied, followed by a Newman–Keuls test of ordered means when appropriate. All hypotheses were tested for significance at the 0.05 level of confidence.

RESULTS

The changes in pulmonary function measured before and after exposure are summarized in Table 3. There was an overall decrease in forced vital capacity under all conditions, including filtered air, with no differences across sulfate concentration. This was apparently the result of a modest decline in inspiratory capacity. The decreases in vital capacity and inspiratory capacity averaged 106 and 121 ml, respectively, which are within the daily variation of these measurements. The only measurement to show an interaction across time (pre–post) and sulfate concentration was $FEV_{1.0}$, which was significantly decreased with exposure to 939 $\mu g/m^3$ sulfate, but not at the other concentrations. However, the average decrease was only 101 ml. None of the other measures showed significant changes during the sulfate exposure.

TABLE 2
PHYSICAL CHARACTERISTICS OF THE EXPOSURE ENVIRONMENT

		Pollutant level ($\mu g/m^3$ SO_4)		
	Filtered air	223	418	939
Sulfate ($\mu g/m^3$)	N.D.[a]	223	418	939
Mass median particle diameter (μm)	0.30	0.93	0.91	0.91
Geometric SD	1.65	1.73	1.71	1.66
Temperature (°C)	22.8	22.5	21.9	22.0
Relative humidity (%)	51	57.3	53.6	52.7

[a] Not detectable.

TABLE 3
PULMONARY FUNCTION MEASUREMENTS IN SUBJECTS EXPOSED TO VARIOUS CONCENTRATIONS OF SULFURIC ACID

		Sulfuric acid ($\mu g/m^3$)			
		0	223	418	939
Forced vital capacity (liters)	Pre	5.64 (±0.58)	5.65 (±0.55)	5.66 (±0.61)	5.65 (±0.61)
	Post[a]	5.58 (±0.56)	5.54 (±0.63)	5.59 (±0.63)	5.47 (±0.60)
$FEV_{1.0}$ (liters)	Pre	4.47 (±0.46)	4.83 (±0.54)	4.72 (±0.55)	4.82 (±0.57)
	Post[b]	4.79 (±0.48)	4.80 (±0.57)	4.76 (±0.55)	4.72 (±0.57)
$FEV_{3.0}$ (liters)	Pre	5.58 (±0.59)	5.61 (±0.57)	5.61 (±0.62)	5.62 (±0.63)
	Post[a]	5.55 (±0.57)	5.51 (±0.64)	5.54 (±0.63)	5.44 (±0.57)
Inspiratory capacity (liters)	Pre	3.62 (±0.48)	3.58 (±0.49)	3.61 (±0.55)	3.54 (±0.50)
	Post[a]	3.48 (±0.45)	3.49 (±0.58)	3.49 (±0.46)	3.40 (±0.46)
Expiratory reserve volume (liters)	Pre	2.02 (±0.45)	2.02 (±0.36)	2.05 (±0.47)	2.12 (±0.39)
	Post	2.10 (±0.45)	2.05 (±0.38)	2.12 (±0.45)	2.06 (±0.40)
Functional residual capacity (liters)	Pre	3.66 (±0.74)	3.68 (±0.73)	3.72 (±0.69)	3.74 (±0.74)
	Post	3.73 (±0.72)	3.76 (±0.70)	3.80 (±0.76)	3.76 (±0.73)
FEF 25–75% (liters/sec)	Pre	4.97 (±0.69)	5.14 (±0.89)	4.97 (±0.74)	5.11 (±0.88)
	Post	5.06 (±0.81)	5.36 (±1.11)	5.05 (±0.80)	5.29 (±1.08)
Max voluntary ventilation (liters/min)	Pre	202 (±34)	206 (±35)	199 (±35)	211 (±35)
	Post	205 (±33)	202 (±41)	204 (±39)	209 (±32)
Airway resistance (liters/sec/cm H_2O)	Pre	1.67 (±0.39)	1.73 (±0.35)	1.75 (±0.52)	1.87 (±0.48)
	Post	1.76 (±0.58)	1.77 (±0.57)	1.64 (±0.46)	1.69 (±0.41)
FEF 50%	Pre	5.57 (±0.77)	5.85 (±1.01)	5.97 (±0.85)	5.82 (±0.97)
	Post	5.63 (±1.03)	6.12 (±0.98)	5.84 (±0.96)	5.81 (±1.15)
FEF 75% (liters/sec)	Pre	2.80 (±0.65)	2.83 (±0.57)	2.83 (±0.68)	2.76 (±0.61)
	Post	2.93 (±0.80)	3.05 (±0.90)	2.88 (±0.77)	2.89 (±0.73)
FEF 75–85% (liters/sec)	Pre	2.12 (±0.59)	2.20 (±0.54)	2.29 (±0.61)	2.13 (±0.68)
	Post	2.22 (±1.02)	2.32 (±0.72)	2.32 (±0.70)	2.28 (±0.71)

Note. Pre and post 2-hr exposures; mean ± SD.
[a] Post < pre overall—no difference across sulfate concentrations.
[b] Pre (FA, 418) < pre (223, 939); pre (939) > post (939).

Table 4 presents pulmonary function data obtained immediately (2–3 min) after the subjects had completed each of their three exercise bouts where pulmonary ventilation had been at a level of 30 liters/min for the previous 20 min. At this level of ventilatory exchange subjects are still predominantly nasal breathers (D'Alfonso, 1980; Niinimaa *et al.*, 1980). Despite the increased intake of sulfuric acid aerosols, no significant changes were observed in any of the functions measured.

Symptoms

Symptoms reported by the subjects following the exposure are summarized in Table 5. Many subjects noticed a dry or metallic taste in the air. At the highest concentration, throat irritation or dryness and cough were frequently noted. Eye

TABLE 4
PULMONARY FUNCTION MEASUREMENTS DURING 2-hr EXPOSURE TO SULFURIC ACID

		Sulfuric acid ($\mu g/m^3$)			
	Time (min)	0	223	418	939
Forced vital capacity (liters)	Pre	5.35 (±0.54)	5.25 (±0.55)	5.30 (±0.54)	5.28 (±0.59)
	25	5.21 (±0.61)	5.25 (±0.56)	5.20 (±0.58)	5.20 (±0.59)
	65	5.24 (±0.62)	5.21 (±0.56)	5.24 (±0.56)	5.17 (±0.61)
	105	5.16 (±0.62)	5.23 (±0.57)	5.17 (±0.53)	5.13 (±0.62)
$FEV_{1.0}$ (liters)	Pre	4.65 (±0.49)	4.53 (±0.51)	4.51 (±0.57)	4.55 (±0.57)
	25	4.48 (±0.54)	4.43 (±0.64)	4.43 (±0.59)	4.49 (±0.55)
	65	4.53 (±0.58)	4.45 (±0.53)	4.52 (±0.49)	4.44 (±0.55)
	105	4.52 (±0.52)	4.56 (±0.64)	4.53 (±0.53)	4.48 (±0.57)
$FEV_{3.0}$ (liters)	Pre	5.33 (±0.56)	5.22 (±0.56)	5.28 (±0.55)	5.26 (±0.59)
	25	5.18 (±0.61)	5.19 (±0.60)	5.12 (±0.59)	5.16 (±0.59)
	65	5.23 (±0.63)	5.17 (±0.59)	5.19 (±0.56)	5.15 (±0.62)
	105	5.13 (±0.61)	5.20 (±0.54)	5.14 (±0.54)	5.10 (±0.20)
Inspiratory capacity (liters)	Pre	3.48 (±0.48)	3.39 (±0 57)	3.43 (±0.43)	3.43 (±0.54)
	25	3.25 (±0.59)	3.23 (±0.58)	3.34 (±0.49)	3.29 (±0.46)
	65	3.29 (±0.43)	3.32 (±0.55)	3.31 (±0.49)	3.21 (±0.49)
	105	3.19 (±0.42)	3.36 (±0.60)	3.26 (±0.45)	3.22 (±0.48)
FEF 25–75% (liters/sec)	Pre	4.92 (±0.79)	4.91 (±0.83)	4.75 (±0.97)	4.90 (±0.83)
	25	4.73 (±0.83)	4.82 (±1.21)	4.64 (±0.97)	4.90 (±0.84)
	65	4.90 (±1.01)	5.13 (±1.05)	4.81 (±0.89)	4.84 (±0.95)
	105	5.05 (±0.83)	5.10 (±1.30)	4.90 (±0.89)	5.02 (±0.80)
FEF 50% (liters/sec)	Pre	4.80 (±0.83)	4.99 (±0.78)	4.69 (±0.99)	4.92 (±0.85)
	25	4.73 (±0.81)	4.76 (±1.14)	4.54 (±0.94)	5.00 (±0.86)
	65	4.88 (±0.98)	4.98 (±0.84)	4.76 (±0.91)	4.92 (±0.89)
	105	5.05 (±0.80)	5.03 (±1.08)	4.83 (±0.93)	5.00 (±0.92)
FEF 75% (liters/sec)	Pre	2.53 (±0.61)	2.66 (±0.70)	2.40 (±0.74)	2.54 (±0.58)
	25	2.51 (±0.71)	2.50 (±0.65)	2.49 (±0.84)	2.56 (±0.57)
	65	2.53 (±0.84)	2.64 (±0.77)	2.47 (±0.72)	2.52 (±0.68)
	105	2.65 (±0.73)	2.66 (±0.87)	2.63 (±0.78)	2.59 (±0.66)

Note. Mean ± SD.

irritation was reported by two subjects at 939 $\mu g/m^3$ of sulfate. Several individuals reported dizziness, fatigue, or headache, which were usually associated with exercise and had no relationship with sulfate levels in the ambient air.

DISCUSSION

These data indicate that there is no significant effect of sulfuric acid aerosol in the 1-μm (0.9-μm)-size range and in concentrations between 200 and 1000 $\mu g/m^3$ on the pulmonary function of young male nonsmokers. The results of the present study support previous studies (Avol *et al.*, 1979; Kerr *et al.*, 1981; Sackner *et al.*, 1978; Lippman *et al.*, 1981; Leikauf *et al.*, 1979) of human exposure to sulfuric acid aerosols in the 0.3- to 0.5-μm-size range at similar concentrations (100–1000

TABLE 5
NUMBER OF SUBJECTS REPORTING SYMPTOMS CONSEQUENT TO EXPOSURE

		Pollutant level ($\mu g/m^3$ SO_4)		
	Filtered air	223	418	939
Unusual odor or taste	0	7	3	6
Sore throat, irritation, dryness	1	3	5	8
Cough	0	2	5	8
Shortness of breath, chest tightness	0	3	3	3
Dizziness, fatigue, or headache	6	4	2	6
Eye irritation	0	1	1	2
No symptoms	4	2	2	0

$\mu g/m^3$). However, they do not confirm the studies of Amdur *et al.* (1952) and Sim and Pattle (1957), whose subjects were exposed to larger aerosols, as in our study. We not only exposed our subjects for longer periods but intentionally increased the quantity of inhaled aerosol by having them exercise so that their ventilatory volumes were intermittently increased up to 30 liters/min. Larson and co-workers (1977, 1978) have demonstrated that ammonia, which is present in mouth and pharynx, is capable of neutralizing some of the inhaled sulfuric acid aerosol; the amount neutralized depends on a number of factors, including residence time in the upper airway, H_2SO_4 content of the aerosol, particle size of the aerosol, and relative humidity. Such a neutralization process may be partially responsible for the lack of effect of the sulfuric acid aerosol.

Higher H_2SO_4 concentrations (2.4–5.0 mg/m^3) and prolonged exposure (78 weeks) have produced deleterious changes in both lung function and lung morphology in cynomolgus monkeys (Alarie *et al.*, 1973).

The increased prevalence of symptoms such as cough, dry throat, or throat irritation suggests that the sulfuric acid may have caused some irritation to the tracheal mucosa although not to a sufficient degree to induce functional changes. These symptoms were usually noted only when the subject took a deep or rapid inspiration and generally only at the beginning of the exposure period.

Our subjects were young, healthy individuals who have been living in a low-pollutant environment. Consequently, care should be taken in extrapolating these results to individuals who may have varying degrees of pulmonary dysfunction.

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